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CHARACTERIZATION OF THERMALLY DAMAGED LX-17

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Characterization of Thermally-Damaged LX-17

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Abstract

Thermal damage was applied to LX-17 at 190 ^{o}C for several hours. The damaged LX-17 samples, after cooled down to room temperature, were characterized for their material properties (density, porosity, permeability, moduli), safety, and performance. Weight losses upon thermal exposure were insignificant (< 0.1% wt.). The damaged LX-17 samples expanded, resulting in a bulk density reduction of 4.3%. Subsequent detonation measurements (cylinder tests) were conducted on the thermally-damaged LX-17 samples. The results showed that the fractions of damaged LX-17 reacted were slightly lower than those of pristine LX-17. The thermally damaged LX-17 had a detonation velocity of 7.315 mm/μs, lower than that (7.638 mm/μs) of pristine LX-17. Detonation energy density for the damaged LX-17 was 5.08 kJ/cm^3, about 9.0% lower than the detonation energy density of 5.50 kJ/cm^3 for the pristine LX-17. The break-out curves showed reaction zone lengths for pristine LX-17 and damaged LX-17 were similar but the damaged samples had ragged detonation fronts.

Keywords: TATB, LX-17, Thermal Damage, Cylinder Test, Detonation Velocity, Detonation Energy, Edge Lag

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1. Introduction

Thermal incidents may expose energetic materials (EM) to unexpected heat that may damage the explosive charge (e.g., change microstructure, introduce voids and porosity, and increase surface area). This may affect material properties, sensitivity, safety, and performance of the energetic materials. Reusing the damaged explosives requires a thorough analysis of the materials. Characterization methods and changes in material properties for thermally damaged HMX-based formulations (LX-04 and LX-10) were reported elsewhere by Hsu et. al [1,2]. LX-04 and LX-10 consist of 15% and 5% viton A, respectively and balance of HMX. In general, HMX-based formulations experienced an irreversible volume expansion by more than 5% after thermal damage at temperature above 170 ^{o}C. Both gas permeabilities and burn rates of the damaged samples increased by several orders of magnitude due to higher porosity and lower density. The thermally damaged material also became weaker mechanically and easily to break apart. Some damaged samples were also evaluated at room temperature for their sensitivities to impact, friction and spark (small-scale safety tests). Although no apparent changes in room-temperature sensitivities were found, the materials may be more sensitive to impact at high temperatures. Urtiew et. al reported that heated LX-04 was more sensitive to shock initiation at high temperatures [3].

Other important factor in considering the reuse of damaged material is its detonation performance (detonation velocity and detonation energy). Hsu et. al [4] recently reported that detonation velocity of thermally damaged LX-04 was 7.7 to 7.8 mm/μs, about 10% lower than that (8.5 mm/μs) of pristine high-density LX-04. Detonation energy density for the damaged LX-04 was 6.5 kJ/cm^3, much lower than the detonation energy density of 8.1 kJ/cm^3 for the pristine high density LX-04. The break-out curves for the detonation fronts showed that the damaged LX-04 had longer edge lags than the pristine high density LX-04, indicating that the damaged explosive was less ideal.

LX-17 is a TATB (1,3,5-triamino-2,4,6-tranitrobenzene) -based high explosive developed by Lawrence Livermore National Laboratory (LLNL). It is an insensitive energetic material and is widely used. In this article, we will share our findings on changes in material properties and detonation performance of damaged LX-17.

2. Characterization of damaged energetic materials

Explosive charges in either confined (weaponry, munitions) or unconfined (pressed parts, shape charges, or powders stored in warehouses, magazines, bunkers, ships) environments may suffer unexpected insults which may cause undesirable reactions. Examples of insults are fires, earthquakes, problems associated with operational handling, transportation, sudden climate change, and combat operations in battlefields. Safety/sensitivity data for the damaged materials need to be established if the materials are destined for dismantling and demilitarization, as shown in Figure 1. If stakeholders decide to reuse the damaged explosive charges, a comprehensive characterization of the materials would be desirable, as shown in Figure 2. The High Explosives Applications Facility (HEAF) at Lawrence Livermore National Laboratory is a state-of-the-art facility and is dedicated to energetic materials R&D. It is equipped with many sophisticated devices, equipment, and instruments that offer means for diagnostics and characterization of energetic materials as listed in Table 1. Results of damaged material characterization for LX-04 and LX-10 have been reported elsewhere [1,2,3,4].

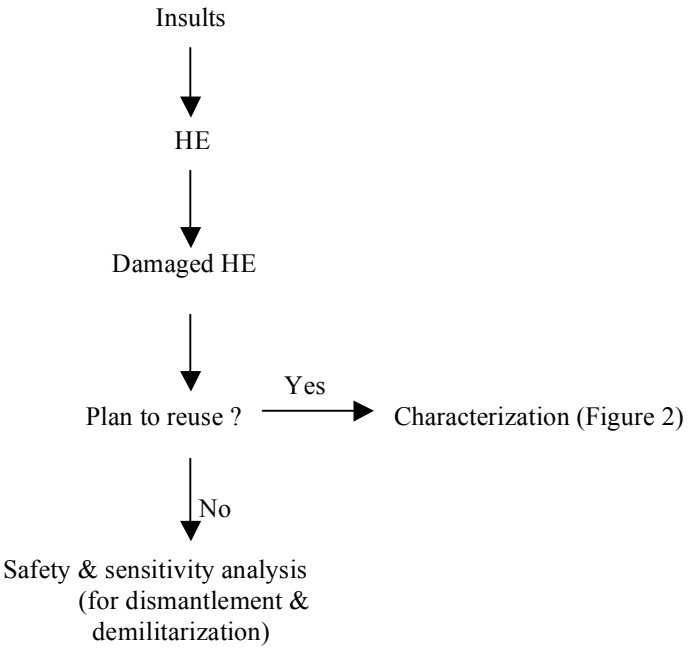


Figure 1. Decision process for the reuse of damaged explosives

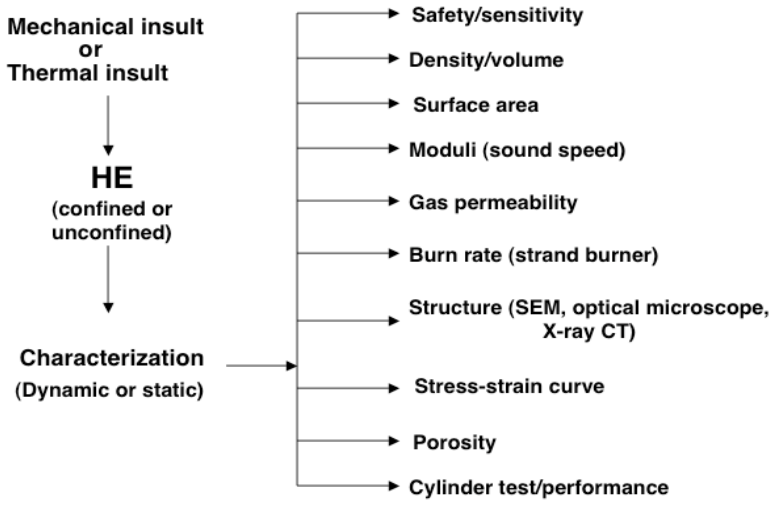


Figure 2. Characterization of damaged energetic materials

Table 1. System and instruments available for damaged material characterization

Instruments/equipment	Measurements
BET	Surface area
Micropycnometer/pycnometer	Volume and density
Gas permeameter	Gas permeability
Strand burner	Burn rate
STEX	Cook off study
Ultrasound probe	Sound speed
Hopkinson bar	Stress-strain curve at high strain rates
Compression equipment, tension equipment	Stress-strain relationship, moduli
Drop weight machine	Impact sensitivity
Friction test machine	Friction sensitivity
Spark test machine	Spark sensitivity
Differential scanning calorimetry	DSC
Scanning electron microscope	Surface structure
Shot tanks up to 10 kg TNT	Shot experiment, cylinder test
One dimensional time to explosion system	Thermal kinetics
4" Gun for high-velocity impacting	Run distance to detonation (Pop-plot)

3. Thermal experiments and results

Thermal damage experiment was conducted remotely in an unconfined environment at high temperature. 12 cylindrical pressed parts of LX-17 (each one was 25.4 mm ϕ x 25.4 mm long) were heated in a 1.0 kg shot tank at 190 ^{o}C for 4 hours. The samples were then cooled and characterized. Weight losses were only 0.09% wt, very insignificant. Sample expanded slightly, results in a bulk density reduction of 4.3%, as shown in Table 2.

Table 2. LX-17 Sample volume and bulk density after thermal damage for each cylindrical part (25.4 mm ϕ x 25.4 mm)

Sample	Wt., g	Bulk volume, cc	Bulk density, g/cc	%TMD*
Pristine LX-17	24.511	12.795	1.916	98.56
Heated at 190 °C, 4 hrs	24.490	13.356	1.834	94.34
% Change	-0.090	+4.4	-4.3	

* TMD (theoretical maximum density) of LX-17 is 1.944 g/cc

Porosity increased after the pressed parts were heated and expanded. Bulk volume of certain sample configurations can be measured accurately by a micrometer and it includes volume occupied by open pores. Blind pores and through pores are open pores which are reachable by gas molecules. The gas pycnometer uses gas displacement principle with a gas pressure of 20 psig and the density obtained from the measurement is called true density. It is very closed to theoretical maximum density (TMD) of the sample if the fraction of closed pores in the sample is insignificant. Total porosity, fraction of closed pores, fraction of open pores can be estimated by Equations 1, 2, and 3. Increase in porosity also resulted in lower mechanical strength (lower moduli), as reported by Hsu et. al. [2].

$$\varepsilon = (TMD - \rho_b)/TMD \quad (1)$$

$$f_c = (TMD - \rho_t)/TMD \quad (2)$$

$$f_o = \varepsilon - f_c \quad (3)$$

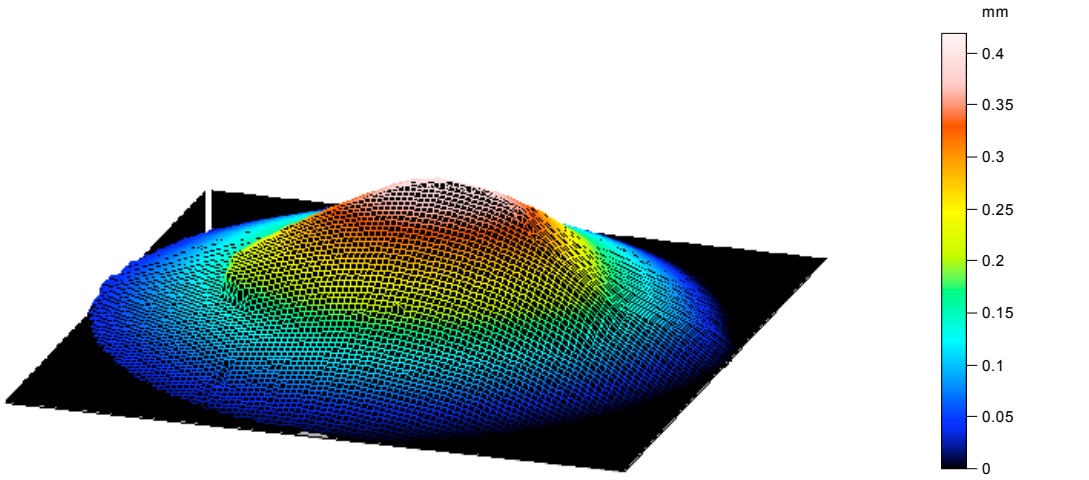
Where ε = total porosity, dimensionless
ρ_b = bulk density, g/cc
ρ_t = true density, g/cc
f_c = fraction of closed pores; dimensionless
f_o = fraction of open pores; dimensionless

Table 3 shows the total porosity, fraction of closed pores, fraction of open pores of LX-17 samples after the thermal exposure for 4 hours at 190 °C. The samples became much more porous, evidenced by the increase of total porosity from 2.0% to 7.4%. Much of the porosity increase came from the open pores (20 times). Similar observation was made on LX-10 [2]. It seems that the use of pycnometer and micrometer can be useful approximation of porosity measurement.

Table 3. Porosity, fraction of closed pores, fraction of open pores of LX-17 samples; samples were cylindrical discs (25.4 mm ϕ x 5.0 mm)

Sample	ρ_b, g/cc	ρ_t, g/cc	ε, %	f_c, %	f_o,%
Pristine LX-17	1.905 98.00% TMD	1.910 98.27% TMD	2.0	1.73	0.27
Damaged LX-17	1.800 92.60% TMD	1.905 97.98% TMD	7.4	2.02	5.38
% Change	- 5.40%	- 0.29%	+5.4	0.29	5.11

A surface profile meter was used to examine surfaces of damaged parts. The cylindrical parts deformed and concaved, as shown in Figure 3. Up to 0.4 mm peak was seen on the cylindrical surface. The data were useful for sizing the copper tube for cylinder shots.



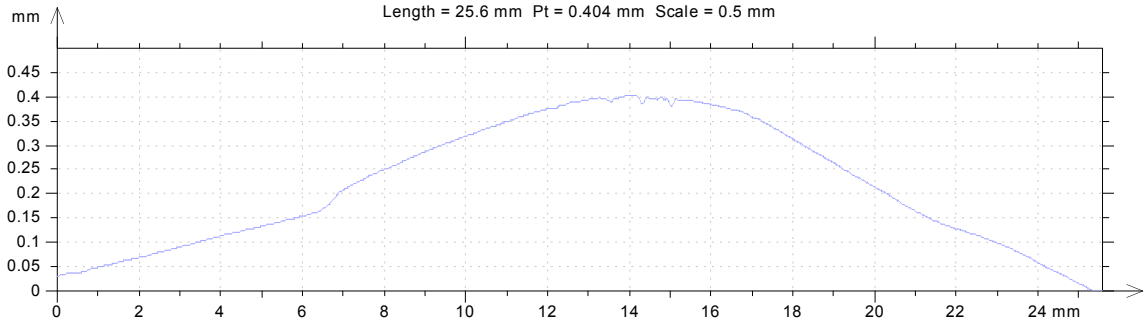


Figure 3: surface profiles of damaged LX-17 samples; profiles shown are for 3D (top) and 2D (bottom)

4. Detonation experiments and results

Twelve damaged LX-17 cylindrical samples were assembled inside a copper tube for cylinder test. The cylinder test is a 40 year-old method for measuring the detonation energy at specific relative volumes of expansion, ranging from about 2 to 7 [5]. The detonation velocity is also obtained from the time difference between pin rings and the breakout curvature of the detonation front is taken from a streak camera looking at light from a slit on the detonation front. A typical cylinder test is shown in Figure 3.

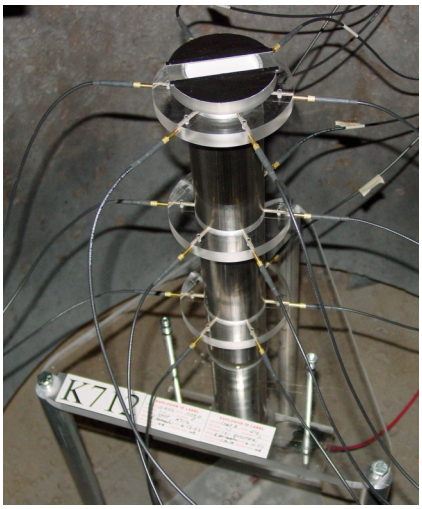


Figure 3. Cylinder test, where the detonation runs upward. The three pins rings are visible as is the end slit for breakout. The laser beam for wall velocity will hit the copper about 2/3rds of the way up. The cylinder sits inside a shrapnel catcher inside the 1.0 kg shot tank.

Figure 4 shows the measured copper wall velocity for the damaged LX-17 shot (#769) as lying just below the historical as-pressed results. Four of the old shots were done by streak camera, and the wide scatter is evident. #628 was done with Fabry interferometry and is directly comparable with #769. The times are "scaled" to 1-inch diameter, which allows the plotting of various sizes of cylinders with different wall thicknesses. However, #628 was a 1-inch diameter shot and #769 is 1.013 inches (because of the swelling), so that the two are directly comparable.

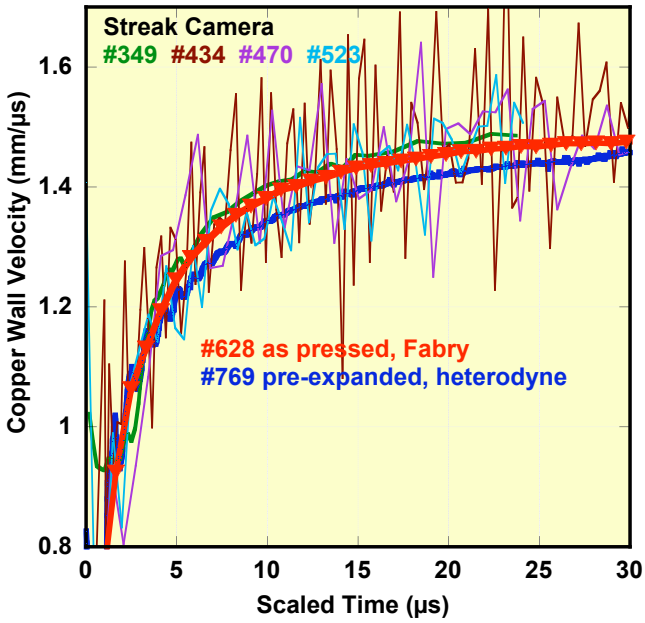


Figure 4. Comparison of copper wall velocities as a function scaled 1-inch diameter times.

The first approach for determining the detonation energy was to equate the square of the copper wall velocity with energies of explosives believed to be near-ideal [6]. Then, the calculated detonation energies

from the thermo-chemical code CHEETAH became good enough to be used as the standards for full burn [7]. The three standard relative volumes were reset to 2.2, 4.4 and 7.2 for scaled wall displacements of 6, 12.5 and 19 mm, respectively. For scaling, a 12.7 mm radius (1 inch diameter) tube was taken as the standard. The copper tunes were roughly divided into types: full-wall (radius equal to $1/5^{th}$ radius) and half-wall ($1/10^{th}$ radius). Full-wall tubes are the present standard because they are cheaper to make. The half-wall was an attempt to get more velocity in the days of the inaccurate streak camera. At LLNL, the streak camera was replaced by Fabry-Perot interferometry, and that has been replaced in-turn by Ted Strand's Hetereodyne Velocimetry [8]. Both laser methods are more accurate than the streak camera, and the Heterodyne is much cheaper than the Fabry. The error bars for streak camera detonation energies are set at ±3%, although some of this is probably material error. The two laser error bars are lower than ±3%.

The measured wall velocities go into a Gurney-type equation which keeps the density of the copper wall constant with expansion [9]. The equation is

$$E_d = \alpha\beta\rho_o\left[\frac{\rho_m}{\rho_o}\left(\frac{R+x}{R_o}\right)^2 \ln\left(\frac{R+x}{R}\right) + \frac{1}{4}\left(\frac{R+x}{R}\right)^2\right]u_m^2. \quad (4)$$

The result is the detonation energy at some given relative volume, which is described with the measured wall velocity u_m. R_o is the initial radius, R the radius at time t and x the wall thickness at time t. The initial densities are r_m for the metal and r_o for the explosive. E_d is the calculated detonation energy and E_d(Ch) will be the detonation energy from CHEETAH. The coefficients a and b are experimental adjustments for the method of measurement, as described above, and the wall type. This equation allows us to successfully calculate early full-wall shots that used metals other than copper and to adjust for small variations in dimensions. The resulting detonation energy, E_d, is then compared with that calculated by the thermo-chemical code CHEETAH and the fraction reacted, F, at any relative volume is given by

$$F = \frac{E_d(\text{measured})}{E_d(\text{CHEETAH})}. \quad (5)$$

It has become traditional to take special points at scaled wall displacements of 6, 12.5 and 19 mm. The displacement is the measured distance that the outer copper wall has moved, which is directly measured by streak camera and obtained by integration from the laser methods. Scaled means that all cylinders are treated as being 12.7 mm inner radius (1 inch inner diameter). Shot #757 was 1.05 times larger than 12.7 mm radius, so that time and displacement (but not velocity) are divided by 1.05 to get the scaled results. Scaling allows comparisons for different geometries. The three displacements mentioned above are correlated to average relative volumes of 2.2, 4.4 and 7.2, respectively. Using the detonation energies at these volumes plus the density and detonation velocity, we can create the JWL Equation-of-State. The Cylinder test and detonation calorimetry are the only methods for obtaining directly-measured detonation energies.

Table 4 shows the results for this shot #769 plus the historical work done on as-pressed LX-17. The detonation velocity, U_s, is smaller as a result of the lower density. If we use the empirical equation

$$U_S(\text{low}) \approx \left(\frac{\rho_{o(\text{low})}}{\rho_{o(\text{high})}}\right)^{2/3} U_S(\text{high}) \quad (6)$$

However, the average fraction reacted is about 0.88 for #769 and about 0.90 for the as-pressed. We take other empirical equations

$$U_S(\text{low}) \approx \left(\frac{E_{d(2.2\text{low})}}{E_d(2.2\text{high})} \right)^{0.37} U_S(\text{high})$$

$$U_S(\text{low}) \approx \left(\frac{E_{d(4.4\text{low})}}{E_d(4.4\text{high})} \right)^{0.39} U_S(\text{high}) \qquad (7)$$

$$U_S(\text{low}) \approx \left(\frac{E_{d(7.2\text{low})}}{E_d(7.2\text{high})} \right)^{0.40} U_S(\text{high})$$

that relates detonation velocity with the detonation energy density at relative volumes of 2.2, 4.4 and 7.2. The results from these equations are

$$\begin{aligned} U_S(\text{density}) &\approx 7.44 \ \ \text{mm}/\mu\text{s} \\ U_S(\text{energy}) &\approx 7.40 \ \ \text{mm}/\mu\text{s} \quad . \\ U_S(\text{measured}) &\approx 7.315 \ \ \text{mm}/\mu\text{s} \end{aligned} \qquad (8)$$

The first number is on the basis of a density change only and the second on an energy density change only. The actual number is lower, so that it appears from this and the fraction reacted that we have lost some of the actual energy by way of degradation in the process of thermal damage.

Figure 5 shows the detonation front breakout. The front is running downward with the center in front and the edges lagging. The pressed explosive has a nice smooth curve but the expanded sample shows that the holes are making for a ragged front. The edge lags are the same, however, which means that, roughly, the reaction zone lengths are the same. The reaction zone is the distance over which energy comes out of the explosive to push the front ahead. This differed from the LX-04 where the edge lags were about twice as large for the expanded samples.

Table 4. Detonation velocities and energies for LX-17.

Shot No.		Expl. Density (g/cc)	Detvel #2 (mm/μs)	Inner Diameter (mm)	Wall Thickness (mm)
769	Pre-Expand	1.834	7.315	25.73	2.540
	average	1.908	7.638	25.415	3.340
349	full density	1.900	7.630	25.414	5.210
470	full density	1.904	7.616	25.417	2.718
471	full density	1.906	7.630	25.407	2.724
522	full density	1.908	7.640	25.414	2.723
432	full density	1.908	7.629	25.424	5.189
554	full density	1.910	7.652	25.415	2.717
439	full density	1.912	7.650	25.412	2.714
523	full density	1.917	7.656	25.417	2.722

Shot	Ed (kJ/cc)			Fraction Reacted		
No.	2.2	4.4	7.2	2.2	4.4	7.2
769	4.27	4.86	5.08	0.89	0.88	0.87
	4.65	5.25	5.50	0.91	0.89	0.89
349	4.62	5.25	5.47	0.91	0.89	0.88
470	4.64	5.08	5.44	0.91	0.86	0.88
471	4.51	5.11	5.43	0.88	0.87	0.88
522	4.54	5.33		0.89	0.90	
432	4.83	5.32	5.52	0.94	0.90	0.89
554	4.82	5.42	5.56	0.94	0.92	0.90
439	4.59	5.27	5.53	0.90	0.89	0.89
523	4.64	5.21	5.58	0.91	0.88	0.90

#769 detvel stev 0.007 mm/μs; length 316.7 mm

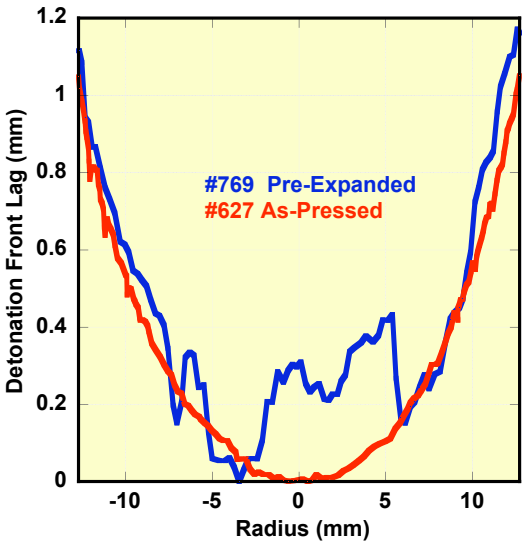


Figure 5. Detonation front breakout curves for LX-17.

Acknowledgments

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